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Building national emission inventories of toxic pollutants in Europe

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ABSTRACT

The reduction of chemical pollution is a priority in many regional, national, and international policies, including in EU countries. To effectively do so, quantified overviews of pollutant emissions at national levels and with some granularity in their sources, are required. However, current monitoring efforts are often scattered and a quantitative and comprehensive inventory of toxic emissions in Europe is lacking. Toxic pollutants stem from a large variety of emission sources from industry, agriculture, households, etc. and the difficulty to cover all of them is manifest in public databases and official reports, where data gaps across countries and years exist for several substances. Here, we propose a methodology to tackle this problem and build comprehensive and harmonized national inventories of toxic pollutants. Using public databases, official reports, scientific literature and developing extrapolation techniques specific to each emission source, we derived harmonized annual inventories of toxic pollutants in all EU Member States over the years 2000–2014. They present an unprecedented coverage of 805, 572, and 468 substances emitted to air, water and soil, respectively. Although the resulting dataset shows a relatively good agreement with previous inventories of narrower scopes, uncertainties can be identified for specific emission sources and in the development of extrapolation techniques, thus calling for further research in these areas. Such efforts should also explore adaptation of the methodology to derive comprehensive inventories for countries outside EU, where data is scarcer. Nonetheless, the developed national inventories can provide a starting point for territorial chemical footprints of toxic pollutants and could be coupled with environmental impact assessment for gauging the damages to ecosystems and human health from toxic pollutants emitted in Europe. This can ultimately support policy-makers in their pollutants prioritisation and benchmarking across substances and countries towards improved toxic emission reduction policies.

1. Introduction

The United Nations' Sustainable Development Goals (UN SDGs) have raised again particular attention on environmental issues such as climate change and pollutant emissions (United Nations, 2015). For example, SDG no. 12, focusing on responsible consumption and production, includes in its targets a significant reduction of the releases of chemicals to air, water, and soil in order to minimize their adverse impacts on human health and the environment (United Nations, 2015). As part of its 2020 Agenda (EC, 2010a), the European Union (EU) is committed to sustainability. The EU is implementing the UN SDGs by integrating them into the European policy framework and evaluating the progress of its Member States (EC, 2016). In this context, it becomes as important to assess the efforts aiming at decoupling the economic growth from the environmental impacts due to the economic activities (UNEP, 2011). This is a key aspect for both the Europe 2020 strategy (EC, 2010a) and its flagship initiative “A resource-efficient Europe”

(EC, 2011), as well as for the Circular Economy Action Plan (EC, 2015).

The thus-necessary monitoring of national emissions of toxic chemicals to air, water and soil is however a complex task hindered by the variety of emission sources (e.g. industrial, residential, agricultural, etc.) and the multitude of existing substances, with for example more than 20,000 substances registered under the ‘Registration, Evaluation, Authorisation and restriction of Chemicals’ (REACH) regulation (EC, 2006a), as reported by the European Chemical Agency (European Chemical Agency, 2018). Inventorying chemicals emissions and characterising their impacts can help measure progress towards the objectives for environmental quality and public health as well as towards a non-toxic environment as envisaged by the 7th Environmental Action Programme (EAP) for EU (EU, 2013; Persson et al., 2019). However, while some initiatives like the European Monitoring and Evaluation Programme (EMEP) support the compilation of pollutant emissions in the EU through national reporting of 25 airborne substances, nationwide inventories often remain limited to a few substances and only

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cover releases of chemicals to water and soil to a limited extent (Cucurachi et al., 2014).

Using Life Cycle Impact Assessment (LCIA) methods, several studies have evidenced the large contribution of water- and soil-borne emissions to total impacts on ecosystems and human health (e.g. Laurent et al., 2011a, 2011b; Lautier et al., 2010; Sala et al., 2015; Sleeswijk et al., 2008). Laurent et al. (2011a) have thus previously estimated that 84% of the toxic impacts on freshwater ecosystems in Europe are caused by emissions of heavy metals to water and soil and by releases of pesticides. This percentage reaches 47% and 50% for toxic impacts on human health, respectively carcinogenic and non-carcinogenic effects, in the same study. Developing national inventories of pollutant emissions with a broad coverage of substances is thus critical to monitor properly their adverse impacts on human health and the environment, as required e.g. for SDGs 3, 15, and 12. While such types of inventories have for example been constructed over time by Sleeswijk et al. (2008) and Cucurachi et al. (2014), they cover only specific years (2000 and 2010 respectively) and do not provide estimates at country level, thus preventing gauging national policies' effectiveness over time.

Here, building on a project initiated in Sala et al. (2014), we aim to tackle this knowledge gap by (i) further developing a methodology for building annual inventories of anthropogenic toxic emissions to air, water, and soil at national scale, and (ii) identifying and defining priorities for future improvements of the inventory. Due to limitations in data availability, the scope of the study is limited to the EU Member States over the period 2000–2014, while the transferability of the methodology to other countries is qualitatively discussed. Because anthropogenic pollutants inducing direct toxic effects on ecosystems and human health are the focus of this study, natural sources of emissions (e.g. volcanic eruptions) and non-toxic substances (e.g. greenhouse gases, nutrients, etc.) are disregarded from our analysis. Despite their possible importance in contributing to impacts, accidental releases are further excluded from this inventory because the focus here is on emissions occurring during normal operations as a result of the current level of policy enforcement and pollution control technologies.

In the current study, we bring significant and novel advances to the work by Sala et al. (2014) and the inventory provided by Sala et al. (2015), including (i) consideration of more emission data with increased substance coverage, (ii) extended temporal and geographic scopes (up to 2014 and for all EU Member States), (iii) use of newly-developed or improved gap-filling procedures and extrapolation techniques, (iv) comparison of the inventories against existing literature, (v) discussion framing the utilisation of the developed inventories, and (vi) provision of national inventories for each year within 2000–2014. With these documented inventories, we therefore intend to provide the means to perform toxic impact assessments at national level in the EU (i.e. part of so-called environmental footprints; Laurent and Owsianiak, 2017), and support benchmarking across EU countries as well as identification of top-contributing substances and associated emission sources that can thereafter be prioritised and tackled through policy-making processes.

2. Material and methods

2.1. Overall methodology

Human economic activities can be categorised into raw material acquisition, incl. agriculture, forestry, fishing and mining (primary sector), construction and manufacturing (secondary sector), and services (tertiary and quaternary sectors) (Kenessey, 1987). Together with households, these human activities exert pressures on the environment and are potential sources of toxic emissions to all environmental compartments.

Industrial activities, such as mining, fuel combustion, metal production, or paper manufacturing, may indeed release heavy metals and/or persistent organic pollutants into air, water, soil compartments

(Fiedler, 2007; Nriagu and Pacyna, 1988; Pacyna and Pacyna, 2001; Tobiszewski and Namieśnik, 2012). In addition, freshwater bodies receive pollutants via domestic or industrial wastewater contaminated with biocides, surfactants, phthalates, polychlorobiphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), and pharmaceuticals (Deblonde et al., 2011). The use of organic waste (including livestock manure and sewage sludge) as fertilizers in agriculture is also known to be a potential source of metal inputs to soil (Alloway, 2013; Belon et al., 2012; Leclerc and Laurent, 2017). While pesticides can be used in specific contexts in industrial or residential settings, they are commonly applied on agricultural crops, where they end up being distributed into air, water and soil media through processes such as drift, runoff, volatilisation, and soil leaching (Dijkman et al., 2012).

Several data sources, such as public databases and literature, document the level of emissions from some of these specific sources, but time series are not always complete and data gaps typically exist for some countries. We thus developed individual methodologies for each emission source; these are summarized in Table 1 and detailed in Sections 2.2–2.5. An overview of the primary data sources and extrapolation procedures used for each emission source is additionally presented in Table A1 in Supporting Information. The toxicity potential and data availability determined the coverage of substances in the present inventory (EC, 2008; UN, 2010). To ensure geographical representativeness, only raw data from EU Member States were used to build the inventories, thus disregarding potential data from countries outside Europe that might generate biases and unjustified substance occurrences in the final inventories. Wherever possible, individual substances were preferred to groups of substances to optimize the potential of the inventories to support impact assessment and targeted decision-making.

2.2. Emissions to air

As illustrated in Fig. 1, airborne emissions of toxic pollutants were mainly based on reports from publicly available national and sectoral databases - like EMEP official reports (EMEP/CEIP, 2017) and the European Pollutant Release and Transfer Register (E-PRTR – Version 11) (EEA, 2017) – see Section 2.2.1. Emissions of non-methane volatile organic compounds (NMVOCs) were however addressed separately (cf. Fig. 1) as they are poorly reported as individual substances in the aforementioned databases (typically as a group; see Section 2.2.2).

2.2.1. Total airborne emissions (excl. NMVOCs)

Total national airborne emissions of 9 metals and 9 persistent organic pollutants were retrieved from EMEP official reports of all EU Member States in the period 2000–2014, except Greece (no data available at the time of the study). Gaps in the time series were preliminary filled by linear interpolation. However, no emission data was reported for Austria, Luxembourg and Slovenia for 6 out of the 9 heavy metals: arsenic, chromium, copper, nickel, selenium, and zinc (metals with voluntary reporting) and thus could not be extrapolated. In the cases where polycyclic aromatic hydrocarbons (PAHs) were only reported as a group, individual emissions of benzo(a)pyrene (B(a)p), benzo(b)fluoranthene (B(b)f), benzo(k)fluoranthene B(k)f, and indeno (1,2,3-cd)pyrene (Indeno) were calculated based on the average mass distribution of PAHs derived from EMEP official reports from all countries in 2000–2014 (i.e. 33.1% B(a)p, 33.3% B(b)f, 16.3% B(k)f, 17.3% Indeno).

To reach a broader substance coverage in the inventory, non-accidental reports of industrial airborne emissions of 38 toxic pollutants (including 2 pesticides that are complementary to agricultural usage addressed in Section 2.5) were retrieved at facility level from the E-PRTR database for all 28 EU Member States in 2001, 2004, and 2007–2015. Although emissions occurring in 2015 are outside the scope of the final inventories (due to incompleteness of inventories for some pollutant sources at the time of this study), these data were

Table 1
Number and types of pollutants included in the national inventories for each emission source and environmental compartment.

Environmental compartment	Emission source	Pollutants covered ^a
Air	Total emissions (excl. non-methane volatile organic compounds, NMVOCs)	9 metals and compounds 15 chlorinated organic substances (incl. dioxins) 14 other organic substances 2 inorganic substances 2 pesticides ^b 270 individual NMVOCs 52 unrefined groups of NMVOCs
	Total emissions of NMVOCs	
Water	Industrial emissions (excl. pharmaceuticals)	22 metals and compounds 25 chlorinated organic substances 35 other organic substances 8 inorganic substances 28 pesticides ^b
	Household emissions (excl. pharmaceuticals)	18 metals and compounds 18 chlorinated organic substances 29 other organic substances 3 inorganic substances 22 pesticides ^b 6 antibiotics 2 estrogen medications 13 other pharmaceuticals
	Total emissions of pharmaceuticals	
Soil	Industrial emissions	8 metals 7 chlorinated organic substances 7 other organic substances 1 inorganic substance 1 pesticide ^b
	Agricultural reuse of manure	8 metals 4 veterinary drugs
	Agricultural use of sewage sludge	7 metals
Air, water & soil	Agricultural use of pesticides	Dioxins (as a group) 173 herbicides 130 insecticides 119 fungicides 16 plant growth regulators 2 other plant protection products

^a List of covered substances available in [Supporting Information B](#), together with full detailed inventories (see [Section 3.1](#)).

^b Pesticides used in a residential or industrial context (as opposed to pesticides used for agriculture, for which the emissions are quantified separately).

retained to increase the robustness of the extrapolation methods (see below).

The reporting to the E-PRTR is limited to industries whose capacities or levels of emissions are above a given threshold. This threshold is specific for each pollutant and sector, and was set so that a complete reporting would cover 90% of the actual total emissions in Europe (EC, 2006b). However, the number of reported pollutant releases differs significantly across countries and receiving environmental compartments, which may indicate that the reporting is inconsistent across EU Member States, and incomplete in countries with a low number of substances reported (Ullrich, 2016). Because emissions of pesticides are strongly regulated at national level in Europe, the absence of reports for

a given country or year was not considered as a data gap but rather as an actual absence of industrial release, and no extrapolation was deemed necessary (i.e. the official E-PRTR reports were directly used in the final inventories). However, it should be noted that there is no direct correspondence between the approval/restriction of a pesticide in a country and the presence/absence of emission reports in that country. For example, there are reports of waterborne emissions of chlordecone in France in 2014, whereas this pesticide was banned in EU under Regulation (EC) No 1107/2009 (EC, 2009a). For other toxic substances, under the assumption of incomplete reporting and since pollutant emissions are highly dependent on the type of industry considered, an extrapolation procedure was developed to fill the data gaps at sectoral level.

As part of this procedure, the Statistical Classification of Economic Activities in the European Community (NACE) was used to aggregate releases into 31 NACE sectors for each pollutant (excl. pesticides), country and year (cf. Table A2). Sectoral gross economic output data, extracted for all EU Member States in 2000–2015 from Eurostat (Eurostat, 2017a), were then used for extrapolations. The sectoral gross economic output was preferred over sectoral gross domestic product (GDP) as it was assumed to be representative of the intensity of the industrial activity in each sector, i.e. the more an industry produces, the higher its economic output is. For each pollutant and sector, emissions normalised by gross economic output were calculated at both national and European levels (expressed in kg of substance emitted per million euro of economic gross output). These were finally combined with country-, year- and sector-specific economic gross output data to extrapolate missing emission reports in each NACE sector. The national emission profile was considered as first choice wherever available; when not possible, the European emission profile was used (see Fig. A1,

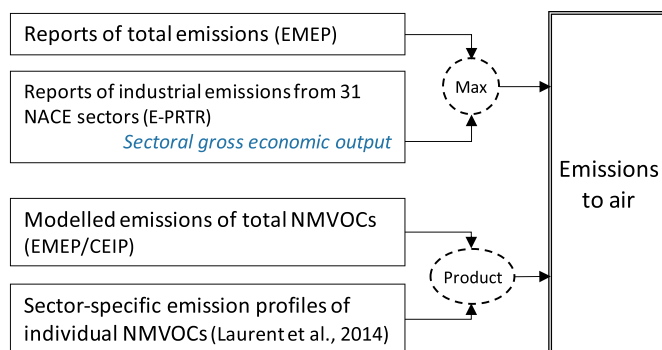


Fig. 1. Methodology developed to estimate nation-wide airborne emissions of toxic pollutants (excl. pesticides) in Europe. Extrapolation proxies are indicated in blue italic. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

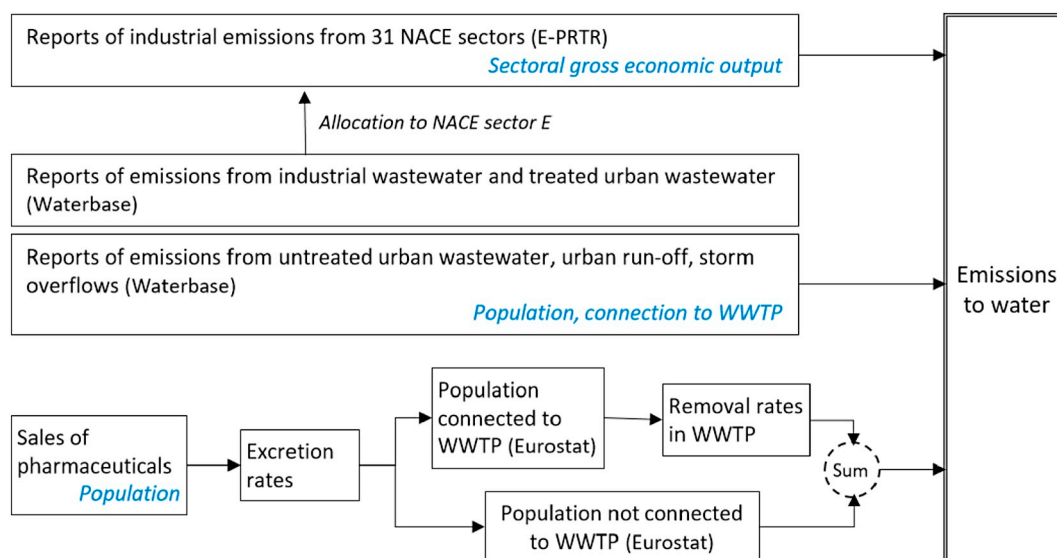


Fig. 2. Methodology developed to estimate nation-wide waterborne emissions of toxic pollutants in Europe. Extrapolation proxies are indicated in blue italic. The treatment of wastewater is considered as an industrial activity covered by the NACE sector E, treated and untreated wastewater are thus considered separately. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

and extrapolation statistics in Supporting Information C). Wherever national emissions of a substance could be estimated from both the EMEP and E-PRTR sources, the highest figure was considered as best estimate due to suspected incompleteness of the reporting in both databases (see Section 3.3.1). Exceptionally, the estimates extrapolated from the E-PRTR were preferred for mercury emissions in Malta as the EMEP official reports are suspected to be highly overestimated (see SI. A).

2.2.2. Airborne emissions of NMVOCs

NMVOCs are reported as a group in both the EMEP official reports (EMEP/CEIP, 2017) and the E-PRTR (version 5 - EEA, 2017). However Laurent and Hauschild (2014) have demonstrated the relevance of considering NMVOCs as individual substances when looking at the toxic impacts on human health and freshwater ecosystems. The methodology developed by Laurent and Hauschild (2014) was adopted to calculate emissions of NMVOCs congeners. By combining speciation profiles retrieved from the literature (i.e. distributions of substances emitted per type of source) and sectoral inventories of total NMVOC emissions (EMEP/CEIP, 2017), Laurent and Hauschild (2014) built country-specific and substance-specific emissions profiles for 107 sectors identified by means of the sector classification NFR09¹ in 2000–2010. In cases where the computations resulted in null emissions of 1,2-dichloroethane, figures were substituted with the available emission data from the E-PRTR database (EEA, 2013).

Since the methodology for NMVOCs breakdown covered only the years 2000–2010 in the original study (Sala et al., 2014), a linear extrapolation at single substance level was performed starting from 2000 to 2010 figures to cover the years 2011–2014. A correction factor was then applied for each year, defined as the yearly increase (or decrease) over the total NMVOC emissions, the latter being retrieved from EMEP/CEIP (2017).

2.3. Emissions to water

As shown in Fig. 2, industrial and residential emissions of toxic pollutants to water were addressed separately due to the scope of the

reporting databases, respectively E-PRTR and Waterbase (EEA, 2017, 2016) – see Sections 2.3.1 and 2.3.2. In parallel, a specific methodology was developed to estimate waterborne emissions of pharmaceuticals (Section 2.3.3), as there is to date little data available for these pollutants of emerging concern (Carvalho et al., 2015).

2.3.1. Industrial waterborne emissions (excl. pharmaceuticals)

Reports of non-accidental emissions of 67 pollutants to water were retrieved at facility level from the E-PRTR database (version 11) for all EU Member States countries in 2001, 2004, and 2007–2015 (EEA, 2017). Pollutant releases due to the discharges of industrial wastewater and treated urban wastewater from non-EPRTR facilities were additionally retrieved from the Waterbase database, version 6 – September 2017 (EEA, 2016). In contrast to Sala et al. (2014), the latter data source was used to complement the E-PRTR reports for the sector NACE E (Water supply, sewerage, waste management and remediation activities) for the relevant pollutant, country and year.

Due to several data gaps remaining in terms of countries and years, extrapolations were required. As with airborne emissions, missing emission data were extrapolated at sectoral level based on the economic gross output for all toxic pollutants except pesticides (see Section 2.2.2, and extrapolation statistics in Supporting Information C). Reports for 2015 were retained to increase the robustness of the extrapolations. Country-, substance-, and year-specific inventories were finally obtained by aggregating the corresponding emissions over all sectors.

2.3.2. Direct emissions from households (excl. pharmaceuticals)

Direct waterborne emissions from households are releases via urban untreated wastewater, unconnected dwellings, urban run-off and storm overflows (emissions related to the treatment of wastewater are addressed together with the industrial sector NACE E, cf. Section 2.3.1). Direct emissions of 114 toxic substances from households were thus retrieved from the Waterbase for 15 EU Member States in 2000–2014 (EEA, 2016). However, several data gaps were observed in terms of substances, countries and years, with for example only six countries reporting emissions from urban run-off and storm overflows. Extrapolations were therefore required to fill the data gaps (Fig. 2).

Demographic statistics (i.e. population count) were assumed the most representative of the magnitude of the residential emissions of pollutants, and were used as extrapolation proxies. Data were retrieved from Eurostat, OECD and The World Bank for all EU Member States in

¹ NFR09 is a Nomenclature for Reporting (NFR) format, i.e. sector classification commonly used in Europe (United Nations Economic Commission for Europe (UNECE)/European Monitoring and Evaluation Programme (EMEP))

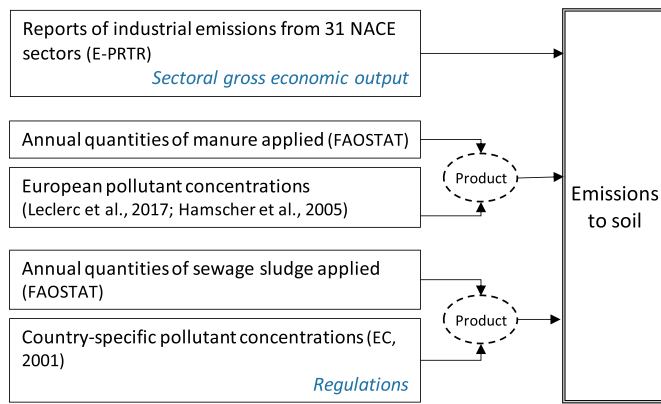


Fig. 3. Methodology developed to estimate nation-wide soilborne emissions of toxic pollutants in Europe. Extrapolation proxies are indicated in blue italic. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

2000–2014 (Eurostat, 2017b, 2017c; OECD, 2017; The World Bank, 2017)). The share of population connected to sewage but without treatment was used as proxy for discharges of untreated urban wastewater, while the share of population not connected to sewage was used as proxy for emissions from unconnected dwellings. Total urban population was used as proxy for pollutant emissions due to storm overflow and urban run-off (see Supporting Information A, and extrapolation statistics in Supporting Information C).

2.3.3. Waterborne emissions of pharmaceuticals

Only emissions of 9 pharmaceuticals are documented in the Waterbase reports from EU countries (EEA, 2016), all stemming from Belgium and the Netherlands in 2013–2014 and 2009–2011, respectively. Considering the paucity of the reported data regarding the emissions of pharmaceuticals to freshwater, it was preferred to derive them from national sales data, taking into account their partial elimination in the human body and removal in wastewater treatment plants (WWTPs). Following Sala et al. (2014), national sales (expressed in kg) of 21 pharmaceuticals were retrieved from the literature for 9 European countries in selected years, and were extrapolated for each Member State and year based on average per-capita sales for each substance (cf. Supporting Information A). These pharmaceuticals are among the ones the most investigated in WWTPs due to their high consumption volumes and/or high toxicity potential (Miège et al., 2009). Sold quantities were assumed to equal the quantities consumed in the respective countries and years, thus disregarding any influence from stock use, carry-over, and imports/exports. Improving the methodology from Sala et al. (2014), Typical substance-specific excretion rates were retrieved from the literature to model the elimination of the pharmaceuticals in the human body and calculate the quantities of chemicals reaching wastewater (cf. Table A5). A distinction between wastewater being treated in WWTPs or not was additionally considered when estimating the emissions (see details in Supporting Information A). Substance-specific removal rates ranging from 0% to 93% were retrieved from the literature, regardless of the type of wastewater treatment (Sala et al., 2014). In the absence of country-specific removal rates, the EU average rate was used.

2.4. Emissions to soil

Inventories of industrial soilborne emissions were built on facility reports from the E-PRTR, whereas agricultural soilborne emissions due to the application of organic waste (manure and sewage sludge) were derived from the respective quantities of manure and treated sludge applied on crops (cf. Fig. 3).

2.4.1. Industrial soilborne emissions

Industrial soilborne releases of 24 toxic pollutants could be extracted from the E-PRTR database (version 11) for 9 EU Member States over the period 2007–2015 (EEA, 2017). As for waterborne emissions, the limited number of countries reporting releases of pollutants to soil indicates that the coverage of the emissions reports for this environmental compartment is likely to be much lower than the claimed target of 90% coverage in the E-PRTR database. In line with industrial emissions to air and water, missing data were extrapolated at sectoral level using the economic gross output as proxy for all toxic pollutants except pesticides (see details in Section 2.2.1 and extrapolation statistics in SI-C).

2.4.2. Agricultural emissions: manure application

Livestock manure contains traces of the chemicals to which the animals have been exposed (e.g. metals, antibiotics, etc.), which are then released to agricultural soil when the manure is used as fertilizer. Emissions of 8 heavy metals were extracted from Leclerc and Laurent (2017) for all EU Member States in the period 2000–2014. The published national inventories are derived from the quantities of manure produced by 16 livestock, assuming that all were used for agricultural purposes. The country-specific shares of livestock manure being managed as solid manure or liquid slurry were retrieved from a survey conducted in 17 European countries (Kuczyński et al., 2005), and the geometric mean was assumed representative of the missing EU Member States. Following a review of measured concentrations of pollutants in manure in European countries between 2001 and 2007, typical livestock-specific pollutant concentrations were calculated. Following the same methodology, additional emissions of 4 veterinary drugs (tetracycline, chlortetracycline, sulfamethazine, and sulfadiazine) were estimated in the current inventory, by averaging the concentrations measured by Hamscher et al. (2005) in Germany in 2001–2003.

2.4.3. Agricultural emissions: sewage sludge application

Similarly to the soilborne releases of pollutants due to the application of manure, emissions of heavy metals from the use of sewage sludge on agricultural land were estimated by combining the quantities of sludge applied and typical concentrations of pollutants. Statistics on the use of sewage sludge in agriculture were retrieved from Eurostat (Eurostat, 2017d) and the United Nations (UN-HABITAT, 2008). Remaining data gaps were filled by linear interpolation between the borders of the gaps when possible, and linear extrapolation over the period 2000–2014 elsewhere (cf. SI-C). Typical concentrations of 7 heavy metals collected at country level in 2005–2006 by the EU Commission (EC, 2010b) were used, assuming their representativeness for the entire time series.

Pollutant concentration profiles were missing for a number of countries, which altogether represented 4% of the total reuse of sewage sludge in EU in 2014. To fill these gaps, it was assumed that countries with similar regulations also shared similar pollutant profiles. Regulations on the thresholds of pollutants from sewage sludge applied to land were thus reviewed (EC, 2010b). As a result, pollutant concentrations of Austria were modelled as those for Germany, Croatia as for Belgium, Denmark as for Slovenia, Greece and Luxembourg as for Cyprus, Ireland as for Spain, and Romania as for Poland (cf. Supporting Information A). Concentrations of organics in sewage sludge are more difficult to obtain and are usually reported as groups of substances (Sala et al., 2014). Due to the low availability of data, only dioxin emissions were included, using an average concentration of 20 ng/kg-dry-matter (EC, 2001).

2.5. Pesticides emissions to air, water and soil

Readily available data on pesticide emissions does not exist; due to confidentiality, even usage or application data are difficult to retrieve at national level. As illustrated in Fig. 4, national applications of pesticides

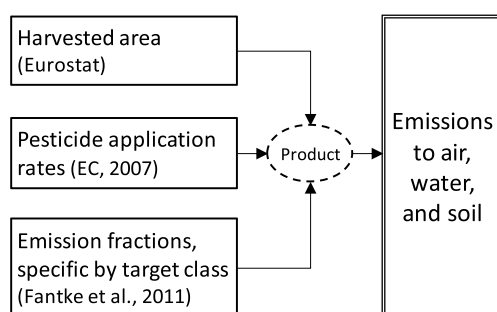


Fig. 4. Methodology developed to estimate agricultural emissions of pesticides to air, water and soil in Europe (simplified from the framework application in Sala et al., 2014).

were calculated by combining harvested area of 9 types of crops (Eurostat, 2017e) and typical pesticide use by crop and country (EC, 2007). Data on pesticide use were compiled for the period 1992–2003, and it is assumed here that they are representative for the entire time series despite potential changes in pesticide authorisations/bans. No data on pesticide use were available for Bulgaria, Romania and Croatia, and due to the large variations in national regulations, no assumption or extrapolation was made to fill these gaps. Whereas pesticide emissions have often been considered as 100% of the applied quantities emitted to soil compartment (like in life cycle inventory databases), distribution processes such as deposition on plants, volatilisation or runoff, occur in the field, meaning that pesticides actually end up in all environmental compartments (Dijkman et al., 2018, 2012). Application data for pesticides should therefore be corrected to account for such partitioning and possible degradation following their application when estimating emissions (Rosenbaum et al., 2015). Assuming a foliar spray

application and the absence of pesticide metabolism in the plant, the dynamic multicrop model dynamiCROP was thus used to calculate average fractions of pesticides reaching air, water surface, soil surface and the plant surface for the main target classes (Fantke et al., 2011). For herbicides, fractions of 10% to plant surface, 15% to air, 74% to soil surface and 1% to surface water were thus considered. For all other target classes of pesticides (insecticides, fungicides, etc.), fractions of 64% to plant surface, 15% to air, 20% to soil surface, and 1% to surface water were considered (Sala et al., 2014).

2.6. Comparison of inventory results

To benchmark the developed inventories and compare the release estimates, studies providing readily available inventories of toxic emissions in Europe in the period 2000–2014, albeit with limited scoping as to substance, year and country coverage, were retrieved from the literature (Table 2). These include the existing work by Sleurswijk et al. (2008), who aimed at estimating total toxic impacts in total Europe in 2000 and as such, their inventory has a large coverage of substances emitted to all environmental compartments. Cucurachi et al. (2014) built a similar type of inventory for the year 2010 but some issues in its documentation prevented its use for comparison (see Supporting Information A). In addition, five other studies were found to focus on the releases of specific toxic substances emitted to air; they were retained for separate analyses with matching substances in our inventory (see Table 2). In the absence of uncertainty quantification of the raw data used in this study, these comparisons can be used as a means to evaluate the precision of the developed estimates, provided that the scopes of both inventories are equivalent.

Table 2

Readily available national inventories of toxic releases retrieved for comparing emission estimates.

Reference	Substance coverage	Temporal coverage	Geographical coverage	Inventory methodology
Sleurswijk et al. (2008)	313 substances to air, water and soil	2000	EU25 + NO, IS, CH ^a (total)	Emissions to air and freshwater are based on reported data from Europe (EMEP, E-PRTR) and US, CA, JA, AU ^a , and extrapolated with national GDP. Metals to industrial soil are derived from Dutch landfill percolate metal loads. Metals to natural soil from manure application are based on livestock statistics, production rates and Dutch metal content in manure. Pesticide use are derived from consumption data in NL, GB, US ^a , and extrapolated based on crop production area for insecticides, and based on GDP for herbicides, fungicides and bactericides.
Breivik et al. (2016)	22 PCB congeners to air	2000–2014	EU (differentiated at country level)	Inventory of PCB emissions (including accidental releases) resulting from the intentional production, use and disposal of PCB-containing products. Mass balance approach including exports of electronic waste, combined with emission factors differentiated by usage/sector and temperature.
Zhang and Tao (2009)	16 individual PAHs to air	2004	World (differentiated at country level; EU countries considered)	Inventory covering emissions from all activities, including fuel combustion, industrial production, consumer product use, and wild fires. Estimates derived from the combination of activity data and country-specific emission factors.
Pacyna et al. (2007)	6 metals to air	2000, 2010	EU28 (differentiated at country level)	Inventory covering emissions from the combustion of various fuels (oil, coal, gasoline, and waste), the production of iron, steel and cement, and the manufacturing of non-ferrous metals, as well as “other sources”. Activity data from the UN Statistical Yearbook and the PRIMES model. Emission factors from EMEP/CORINAIR Guidebook and national experts.
Muntean et al. (2018)	Mercury to air	2000–2012	EU28 (differentiated at country level)	Gridded inventory from the Emission Database for Global Atmospheric Research version tox2 (EDGARv4.tox2). Total emissions based on activity data, emission factors from official datasets, and control measures information from international data sources for key emission sources, i.e. agricultural waste burning, chlor-alkali, power generation, combustion in industry and residential, cement and glass production, non-ferrous and iron & steel industries, waste incineration and transportation.
Quass et al. (2000)	Total dioxins to air	2000, 2005	17 EU countries	Inventory covering most industrial sectors, road transport and accidental fires. Emissions based on official national estimates, activity data and emission factors from the literature. Projections based on developments regarding abatement measures and sector activity.

^a Country codes follow the standard ISO 3166-1.

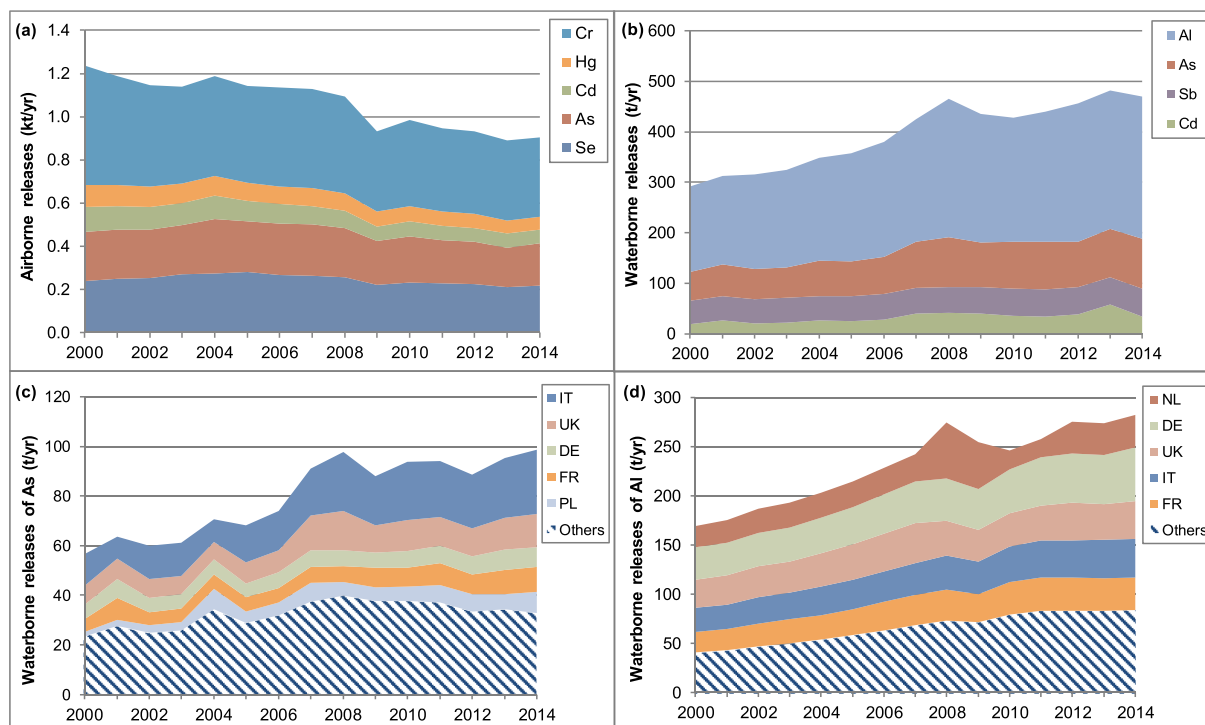


Fig. 5. Illustrative example of trends of selected substance releases (heavy metals). (a) Total airborne releases of 5 selected metals, (b) total waterborne releases of 4 selected metals, (c) distribution of waterborne releases of arsenic between EU countries, (d) distribution of waterborne releases of aluminium between EU countries. Country codes follow the standard ISO 3166-1.

3. Results and discussion

3.1. Resulting national inventories

The newly developed inventories cover pollutant emissions of 805, 572, and 468 substances to air, water and soil, respectively, for each of the 28 EU Member States and for each year within the period 2000–2014 (cf. Table 1 for breakdown overview). The detailed national inventories by emission source are available in Supporting Information B (Excel file). The final inventories cover most of the substances listed under Aarhus protocol on persistent organic pollutants (100%), the EU Water Framework Directive (91%), the guidelines of the World Health Organization (WHO) for drinking-water quality (58%) (EC, 2008; UN, 2010; WHO, 2017). Uncovered substances are either not toxic (e.g. sulfate and sodium in the WHO's guidelines, relevant for water quality but non-toxic), not expected to be used in high volume in EU (e.g. novaluron, insecticide used to control dengue fever), or not associated with emission data at national level (e.g. waterborne emissions of beryllium).

Fig. 5 provides examples of trends observed for selected substances at national and EU levels. These illustrate that the increases or decreases of pollutant emissions in Europe are both substance-specific and country-specific. For example, airborne emissions of several metals tend to decrease at the EU level from 2000 to 2014 (Fig. 5a) while corresponding waterborne emissions tend to increase (Fig. 5b). Likewise, both increases and decreases in metal emissions are observed depending on the metals and the country (see Fig. 5c and d). However, Lim et al. (2010) demonstrated that a quantity-based evaluation of the US Toxics Release Inventory is not sufficient to single out the highest priority chemicals identified with a toxicity-based evaluation. Consequently, to perform further analysis of the contribution of each substance and source and thus support policy-making, impact assessment would be required to account for the relative potencies of each individual substance (Geelen et al., 2009; Laurent and Hauschild, 2014); see discussion in Section 3.5.

3.2. Comparisons with other inventories

3.2.1. Comparison with existing large-scale inventory

For substances present in both our total EU inventory and that of Sleeswijk et al. (2008), estimates of airborne emissions are relatively similar, with ratios ranging from 0.03 to 10.9 (excluding outliers) and a median equal to 0.81 (see Fig. 6). Outliers typically correspond to organic substances for which there is a low level of reporting (e.g. only one E-PRTR report with airborne emissions of ethyl benzene – see SI.C). A larger dispersion of results for emissions to water is observed (Fig. 6); this may be due to the uncertainty associated with the official reports used in both studies (see Section 3.3.1). Emissions to soil are found to be larger (i.e. less than an order of magnitude for most substances) in Sleeswijk et al. (2008) despite a more limited scope of industries (only data from landfill percolate). Although the reporting thresholds in the E-PRTR reports are set to cover 90% of the overall emissions, they may hamper the completeness of the reporting for that emission compartment and thus the accuracy of the results. Among the releases to agricultural soil, a relatively good match is obtained for metals (ratios ranging from 0.2 to 1.7) in spite of releases from the use of sewage sludge not being included in Sleeswijk et al. (2008). However, very large discrepancies can be observed for pesticides because (i) Sleeswijk et al. (2008) assumed that 100% of the pesticides are emitted to soil while this assumption has been refined to 20–70% in this study based on more recent works (see Section 2.5), and (ii) European releases are fully extrapolated in Sleeswijk et al. (2008) whereas country-specific data were used and aggregated to EU level in this study (cf. Table 2).

3.2.2. Comparison to substances-specific inventories

The best fit with bottom-up inventories from literature was obtained for airborne emissions of mercury, with ratios ranging from 0.2 to 4.2 when compared with Pacyna et al. (2007) and Muntean et al. (2018) (see Fig. 7). Overall, the present inventory evaluates the airborne emissions of mercury in EU27 in 2010 to be equal to $6.75\text{E}+04\text{ kg}$, which tends to agree with the estimate of $8.75\text{E}+04\text{ kg}$ done by the

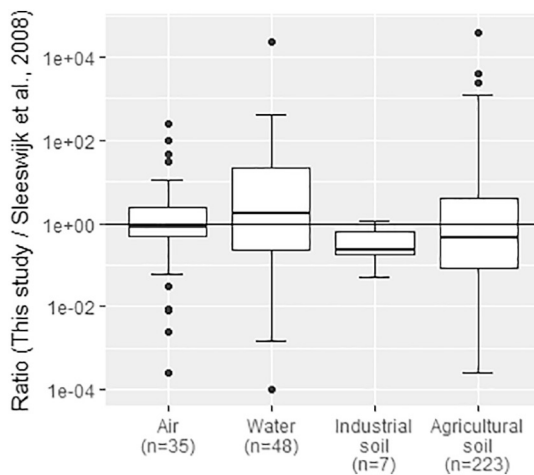


Fig. 6. Comparison of pollutant release estimates with Sleeswijk et al. (2008) for EU-25 + 3 in 2000 (see Table 2). Note the use of logarithmic scale on y-axis. Box plots indicate 25–75 percentile (respectively Q1 and Q3), with the midline indicating the median, while whiskers indicate the minima-maxima excluding outliers. Outliers (dots) are defined as points higher than $Q3 + 1.5IQ$ or lower than $Q1 - 1.5IQ$, with IQ being the inter-quartile range $IQ = Q3 - Q1$.

United Nations (UNEP, 2013). At country level, lower estimates in our inventory were obtained for all other metals in our study, which may indicate that the extrapolations of the E-PRTR reports were not sufficient to balance the potential underestimation of the official EMEP estimates (Pacyna et al., 2007). Furthermore, several outliers can be identified, mostly relating to Malta, Slovenia and Latvia, which may show the limitations of using average European emission factors for all countries as it was done in Pacyna et al. (2007).

The inventory developed in the present study only covers 7 out of the 16 individual PAHs considered by Zhang and Tao (2009), accounting on average for 68% of the total PAHs (expressed in kg B(a)p-equivalent). Emissions of benzo(a)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene, and indeno(1,2,3-cd)pyrene appear to be generally in the same order of magnitude in both inventories (Fig. 7). Those 4 individual PAHs are those prioritised in the reporting to EMEP and E-PRTR (EC, 2006b, 1998), which may explain a consistent reporting. In contrast, emissions of anthracene, naphthalene, and benzo(g,h,i)perylene, which are not covered by the Protocol on Persistent Organic

Pollutants (EC, 1998), appear to be on average an order of magnitude lower in this study than in Zhang and Tao (2009). This apparent underestimation may be due to the lower reporting of these pollutants to the E-PRTR (no EMEP reports available), thus falling outside the scope of the extrapolations performed in our inventory. These findings therefore suggest that additional ways to bypass the reporting thresholds and incompleteness in the reporting are needed to achieve complete national inventories. The complementary use of inventory techniques, combining activity data and emission factors like in Zhang and Tao (2009), could therefore be worth investigating, although consistency should be ensured in such exercise to avoid double counting.

The comparison of our inventories of dioxin emissions to air to the median of the ranges proposed by Quass et al. (2000) shows that both estimates fall within the same order of magnitude, with ratios ranging from 0.29 to 1.2 (excluding outliers). It suggests that the use of official reports allows for an overall good quantification of the releases of dioxins to air.

Lower estimates of total PCB emissions were obtained in this study compared to Breivik et al. (2016), with ratios ranging from $2.6E-04$ to $1.3E+00$ (i.e. spanning 4 orders of magnitude difference). While Breivik et al. (2002) acknowledge that their emission values are at best order of magnitude estimates considering the overall uncertainty of their model, a discrepancy in the scope of both inventories is more likely to explain this difference. First, PCBs are reported as a group to the EMEP and E-PRTR without specification of the individual substances covered, meaning that another set of congeners than in Breivik et al. (2016) may be included. Second, official reports of PCBs, on which our current inventory is built, typically cover non-accidental industrial emissions related to combustion processes or the production and disposal of PCB-containing materials. In those reports, the accounting of emissions during usage of PCB-containing products is often hindered by the lack of methods to quantify them. In contrast, the inventory developed by Breivik et al. (2016) focuses on emissions (accidental or not) due to the production, use and disposal of PCB-containing materials, thus excluding PCB emissions that are occurring as a by-product of other activities such as combustion processes (Breivik et al., 2002). When cumulated over the period 1970–2005, 82% of these global emissions are caused by accidental releases and the usage of PCB-containing products. This analysis suggests that there may be a large gap between the emission coverage of official reports and the actual emissions of PCB including from usage of PCB-containing materials.

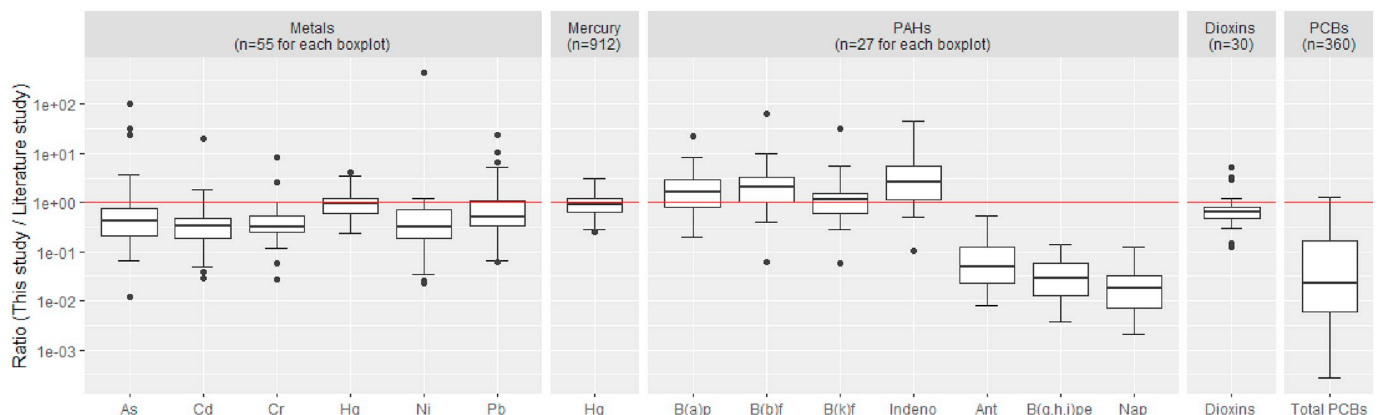


Fig. 7. Comparison of airborne emission estimates obtained in this study with those from literature sources: (i) Pacyna et al. (2007) for metals (individual EU countries, years 2000 and 2010), (ii) Muntean et al., (2018) for Mercury (individual EU countries in 2000–2012), (iii) Zhang and Tao. (2009) for PAHs (individual EU27 countries in year 2004; only 7 out of 16 substances listed in both inventories), (iv) Breivik et al. (2002) for PCBs (individual EU countries in each year of 2000–2014), and (v) Quass et al. (2000) for dioxins (17 EU countries in years 2000 and 2005). Box plots indicate 25–75 percentile (respectively Q1 and Q3), with the midline indicating the median, while whiskers indicate the minima-maxima excluding outliers. Outliers (dots) are defined as points higher than $Q3 + 1.5IQ$ or lower than $Q1 - 1.5IQ$, with IQ being the inter-quartile range $IQ = Q3 - Q1$. As: arsenic, Cd: cadmium, Cr: chromium, Hg: mercury, Ni: nickel, Pb: lead, B(a)p: benzo(a)pyrene, B(b)f: benzo(b)fluoranthene, B(k)f: benzo(k)fluoranthene, Indeno: indeno(1,2,3-cd)pyrene, Ant: anthracene, B(g,h,i)pe: benzo(g,h,i)perylene, Nap: Naphthalene. Note the use of logarithmic scale on y-axis.

Although the comparisons with existing literature remain restricted in their scopes due to a limited number of past studies, they overall tend to show a relatively good agreement. These should be nuanced by important variations when moving from one substance (or group of substances) to another, with some good accuracy in airborne emissions of some metals and organics while showing large gaps for others like PCBs. As exemplified by this latter case, emissions occurring during the use of consumer products can be identified as an important underestimated source of pollutants, which should be addressed in further research.

3.3. Uncertainties and limitations

The inventorying methodologies developed in our study can be categorised into two main types: those mainly relying on public official emission reports (e.g. airborne and industrial emissions), and those building on modelling of emissions from the consumption of products containing toxic pollutants (e.g. the consumption of pharmaceuticals and the use of organic waste for agricultural purposes). In both cases, extrapolations were necessary to build full time series of pollutant emissions in every EU Member States. Sections 3.3.1–3.3.2 discuss the uncertainties specific to these two methodology types, while Section 3.3.3 addresses the robustness of the extrapolation procedures.

3.3.1. Reliability of official emission reports

The inventories of industrial and residential emissions to all environmental compartments were built on three emission-reporting databases, namely the EMEP official reports, the E-PRTR, and the Waterbase. While none of these databases provide quantitative nor qualitative uncertainty estimates at substance level, they appear incomplete in terms of source coverage, and with gaps and inconsistencies in reporting across countries – see the example of mercury emissions to air in Malta in Supporting Information A (Breivik et al., 2006; Manfred et al., 2012; Pacyna et al., 2007; Poupa et al., 2014; Ullrich, 2016). De Marchi and Hamilton (2006) suggest comparing self-reported data with monitored data to assess the overall accuracy of the reporting for a chemical, but such method does not allow identifying which report is inaccurate and requires an extensive monitoring programme at national level. In the absence of sufficient information on the accuracy of the raw data used in its inventory, Pacyna et al. (2007) also compared air concentrations modelled from their releases of metals with measured air concentrations to evaluate the overall accuracy of their estimates. The application of such method is however limited to airborne emissions, for which national measurements exist. Hence it is of limited relevance in the current study to evaluate overall uncertainty due to the large substance coverage in our inventories and the paucity of pollutant emission measurements at national scale.

In practice, despite review procedures thriving to ensure the quality of the reports, outliers due to inconsistent reporting may remain, impacting the reliability of the datasets and the robustness of the extrapolation procedures (Manfred et al., 2012; Mareckova et al., 2013). These are even more difficult to identify in the case of low reporting of emissions of a given substance from a specific sector. For example, there is only one E-PRTR report of vinyl chloride releases to soil in the entire period 2007–2014, which may result in overestimations when extrapolated to all EU countries and years (see Section 3.3.3). Defining a set of recommended methodologies to measure and/or calculate the pollutant emissions should therefore be considered to support a higher consistency of reporting across facilities and countries. In parallel, clarifying the scope of the reporting may aid improve the completeness of the reporting by helping identify which pollutants are released in each sector.

3.3.2. Accuracy of the modelling of pollutant emissions from product consumption

In the absence of emission reports, substance emissions were

modelled based on statistics on the consumption of specific products: e.g. the use of organic waste for agriculture, the application of pesticides on crops, or the consumption of pharmaceuticals. This modelling required various assumptions and simplifications compared to the complex reality of the emission pathways, which results in uncertainties. For example, the assumption that all manure produced by livestock is used as organic fertilizer means that it does not account for its use for biogas production, resulting in potential overestimations of the final releases (Leclerc and Laurent, 2017). Similarly, it was assumed that all pesticides were applied with a foliar spray, which influences the emission fractions reaching each environmental compartment.

In addition to these modelling simplifications, literature data on the concentrations or uses of toxic substances were used to model final releases of pollutants from the consumption of pharmaceuticals, pesticides, and organic waste. The geographical and temporal representativeness of such literature data are limited by the scope and the methodology that differ across existing studies. The lack of harmonized measurements of metals in manure thus compels the use of averaged European concentrations instead of country-specific ones (Leclerc and Laurent, 2017). The removal of pharmaceuticals in WWTPs is highly dependent on the type of treatment and as such may in practice vary strongly from one country to another (Verlicchi et al., 2012), but typical removal rates were considered by default in the present study, regardless of the level of implementation of various treatment technologies. Similarly, the pesticide data compiled in 2005/2006 do not allow accounting for changes in regulations, bans or substitution of active ingredients, nor do they allow evaluating their uses in some EU Member States, namely those that joined after the release of the reference report used in this study and for which data were not available (Bulgaria, Romania, and Croatia). Consolidation and harmonization of these methodologies, e.g. generating standards or exploring possibilities of conducting meta-analysis, should thus be included in future research. Moreover, several substances (such as endocrine disruptors, nanomaterials, etc.) are still not comprehensively covered by any inventory whereas they are related to increased concerns on their potential harm to the environment and human health.

3.3.3. Robustness of the extrapolation procedures

Data gaps in databases were filled by linear regression or using inter-/extrapolation proxies such as gross economic output and demographic statistics. These however do not allow accounting for national differences in pollution control technologies of industries or treatment efficiencies of WWTPs. Depending on the quality of the already existing data (see Section 3.3.1) and the representativeness of the proxy, the robustness of these extrapolation procedures varies from one methodology to another as well as from one pollutant to another. For example, after normalising industrial waterborne emissions from the NACE sectors C31–C33 by the sectoral gross economic output across all countries and years, the geometric standard deviations reach a value of 2000 for xylenes whereas it is equal to 2.5 for copper and compounds (cf. Supporting Information C). Likewise, among direct residential releases, the dispersion of the emissions normalised by the population is nearly twice as big as for emissions from urban run-off and storm overflows than for emissions from unconnected dwellings and untreated discharges (cf. Supporting Information C). Taking into account the frequency of rain events in each country could thus be investigated as a proxy to improve the quality of the extrapolations. Similarly, statistics on the consumption of various pharmaceutical products types or the frequency of disease types could help refine the extrapolation of the sales of pharmaceuticals across countries and years. Finally, there were cases in which the releases of a specific substance in a given sector were only documented by a few reports from a single country, and then extrapolated over countries and years, resulting in possible overestimations of the emissions or in trends being highly influenced by those of the proxies. For example, the reduction in the industrial gross output following the 2008 financial crisis can have a large influence on

the estimates of industrial emissions for substances with a low level of reporting (see Fig. 5d). Although these remain limited to a few substances in the total inventories, e.g. calcium in Estonia or nitrobenzene in the United Kingdom, future works should explore data availability and potential updates to refine these emission estimations.

Owing to the large variations across substances in the accuracy of their respective emission estimates, it is recommended to refine estimation and extrapolation techniques for substances that cause the largest damages to human health and ecosystems; such prioritisation requires performing impact assessment on the developed inventory –see discussion in Section 3.5.

3.4. Applicability of the methodology outside EU

Although the present inventories were developed for EU28 only, reducing emissions of toxic impacts is a global goal, and as such, it is relevant to quantify them in other countries. A high uncertainty is expected from any attempt to extrapolate emissions in non-EU countries based on EU data due to potentially large differences in regulations and technologies (e.g. pesticide use, pollution control technologies) or lifestyles (e.g. pharmaceutical consumption) (Ecobichon, 2001; Persson et al., 2017). For example, the concentration of metals in manure was found to be generally higher in non-European countries compared to the European average (Leclerc and Laurent, 2017). Persson et al. (2019) also acknowledged that a direct extrapolation based on the economic activity may result in underestimations of the emissions outside the EU. The use of local or regional pollutant data is thus fundamental to calculate country-specific emission estimates, meaning that the applicability of our methodology in countries outside EU is highly dependent on the availability of emission reports and pollutant measurements in these countries.

Pollutant Release and Transfer Registers (PRTRs) are being increasingly implemented across the world with, among other things, support from the OECD task force on PRTRs and the knowledge-sharing platform developed by the United Nations Institute for Training and Research (OECD, 2018; UNITAR, 2018). As a result, while PRTRs already exist in Canada, the United States, Australia, Japan, Mexico, Chile, Honduras, Peru, and Israel, they also have been initiated in countries like South Africa, Brazil, Argentina, Cambodia, Thailand, Egypt, etc. (UNECE, 2016). Depending on the format of their reporting, the present methodology may be adapted to estimate industrial emissions to air, water, and soil in these regions. More refined extrapolation techniques as those encompassed in the current work for EU should also be explored, with needs for proxies that factor in the different regulatory, technological and behavioural patterns. Building on this study on EU, Crenna et al. 2019 have proposed a global inventory of emissions.

3.5. Possible uses of the national inventories

The developed national inventories can support many chemical assessments both in academia and policy contexts. Although they are currently estimated at the country and source levels, the developed inventories could support pollution modelling exercises if spatial disaggregation over a gridded domain is performed. Gridded data of population and gross domestic product may for example be investigated to derive gridded emission estimates (Hodges et al., 2014, 2012). In life cycle assessment (LCA), this new inventory can serve as a basis for calculating updated normalisation references for toxicity-related impact categories for the EU up until 2014, representing the average impact of a person over the year (Hauschild et al., 2018). As such, they can help LCA practitioners check and interpret their results by putting them in perspective with a reference level of toxic impacts in EU.

In a broader context, national inventories generally allow for quantifying territorial impacts by combining the pollutant emissions with the fate and effects of the emitted substances in the environment

(causing damages to human health and ecosystems). Environmentally-extended multi-regional input-output tables (EE-MRIOTs) can also be used to link emission profiles to sectors and account for impacts along the global supply-chain of a country (Tukker et al., 2018) with a consumption perspective. Currently, EE-MRIOTs have a good coverage of impacts related to e.g. climate change, land use, and material and water consumption. However, the assessment of toxic impacts from a consumption perspective remains limited by the lack of national inventories of the relevant pollutants with a sectoral differentiation (Steinmann et al., 2018). The inventories built in this study may therefore serve as a starting point to populate environmentally-extended multi-regional input-output tables (EE-MRIOTs) with emission data for toxic pollutants, although additional research is required to allocate emissions to the various sectors of the EE-MRIOT and ensure that all countries of the table are equally covered (see Section 3.4 for countries outside EU). In an attempt to link the E-PRTR data with the sectors of the EE-MRIOT EXIOBASE, Persson et al. (2019) used for example as allocation key the monetary value of the purchases of products from the chemical industry by other industries.

Furthermore, the newly-developed inventories might support policy makers in pollutant prioritisation and benchmarking efforts, allowing for first screening or comparisons of the most contributing emission sources for each substance by differentiating industries, households and agricultural practices. In such efforts, comparisons across countries could also be envisaged. The inventory could be combined either with LCIA or chemical risk assessment methods, depending on the scope (e.g. aiming at average impacts in LCIA or focusing on the most sensitive species to be protected in risk assessment). Both approaches take into account the toxicity potential of each substance and translate emissions into potential damages to human health and ecosystems, towards the calculation of the so-called “chemical footprint” (Laurent and Owsianiak, 2017; Zijp et al., 2014). As exemplified in the chemical footprint assessed for Europe by Sala and Goralczyk (2013), the inventory of each country could thus be characterised, prioritising chemicals based on both their emission quantities and toxicity scores. Such a footprint represents a comprehensive manner to address chemicals, compared to the more traditional substance-by-substance approach (e.g. the one usually adopted in risk assessment for registering or authorising a substance). This can help prioritise the substances to be tackled by policy interventions, such as efforts under REACH and CLP, and specific legislations on groups of chemicals, like biocides (EC, 2012), pesticides (EC, 2009a), pharmaceuticals² (EC, 2008) or cosmetics (EC, 2009b), as well as assist monitoring of the efficiency and efficacy of reduction measures towards set targets such as the “non-toxic environment” defined by the 7th Environment Action Programme (EU, 2013). Used in combination with footprints addressing other environmental impacts such as climate change, land use, material and water consumption, etc., chemical footprints based on our inventory may thus contribute preventing burden shifting from one environmental problem (e.g. climate change) to another (e.g. chemical pollution). It can thus be instrumental to support targeted policy actions in reducing anthropogenic impacts on the environment, as well as in monitoring progress towards environmental sustainability and the decoupling of economic growth and environmental impacts (UNEP, 2011).

4. Conclusion

By developing methodologies that build upon a comprehensive and complementary use of publicly available databases, this study demonstrates the feasibility of building extensive inventories of toxic

² Article 8c of Directive 2008/105/EC (amended by Directive 2013/39/EU) obliges the European Commission to develop a strategic approach to water pollution from pharmaceutical substances

emissions to all environmental compartments in EU Member States over the entire period 2000–2014. Capturing a wide and unprecedented number of substances, with quantified national releases of 805, 572 and 468 substances to air, water and soil compartments over the period of 2000–2014, the study goes beyond previous attempts at estimating toxic pollutant emissions and offers an overarching and differentiated perspective of pollutant releases at EU level. Where possible, comparisons with previous studies focusing on such individual substances or country or years showed that our estimates are generally valid, although they also highlighted important uncertainty pertaining to the pollutants with low level of reporting (like pesticides). Updated statistics on country-specific pesticides consumption and application as well as enhanced monitoring of pollutant releases at national and industrial levels are critical to improve the quality of the developed national inventories. Additional methodological improvement needs to quantify pollutant releases occurring during the use of products at consumer level have also been identified.

Regardless of those improvement potentials, the proposed national inventories are deemed a relevant starting point for undertaking environmental impact assessment and national chemical footprints, thus quantifying the damages to human health and ecosystems from the territorial releases of toxic pollutants, characterising temporal and geographical trends in Europe, and putting them in perspective with other environmental stressors like e.g. greenhouse gases. Such analysis can thus feed into policy initiatives to target those emissions and their sourcing activities carrying the largest environmental burdens.

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2019.03.077>.

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References

- Alloway, B.J., 2013. Sources of heavy metals and metalloids in soils. In: *Heavy Metals in Soil*. Springer, pp. 11–50. https://doi.org/10.1007/978-94-007-4470-7_2.
- Belon, E., Boisson, M., Deportes, I.Z., Eglin, T.K., Feix, I., Bispo, A.O., Galsomies, L., Leblond, S., Guellier, C.R., 2012. An inventory of trace elements inputs to French agricultural soils. *Sci. Total Environ.* 439, 87–95. <https://doi.org/10.1016/j.scitotenv.2012.09.011>.
- Breivik, K., Sweetman, A., Pacyna, J.M., Jones, K.C., 2002. Towards a Global Historical Emission Inventory for Selected PCB Congeners - a Mass Balance Approach. 1. Global Production and Consumption. *Sci. Total Environ.* 290 (1–3), 181–198. [https://doi.org/10.1016/S0048-9697\(01\)01075-0](https://doi.org/10.1016/S0048-9697(01)01075-0).
- Breivik, K., Vestreng, V., Rozovskaya, O., Pacyna, J.M., 2006. Atmospheric emissions of some POPs in Europe: a discussion of existing inventories and data needs. *Environ. Sci. Pol.* 9, 663–674. <https://doi.org/10.1016/j.envsci.2006.09.001>.
- Breivik, K., Armitage, J.M., Wania, F., Sweetman, A.J., Jones, K.C., 2016. Tracking the global distribution of persistent organic pollutants accounting for E-waste exports to developing regions. *Environ. Sci. Technol.* 50, 798–805. <https://doi.org/10.1021/acs.est.5b04226>.
- Carvalho, R.N., Ceriani, L., Ippolito, A., Lettieri, T., 2015. Development of the First Watch List under the Environmental Quality Standards Directive. <https://doi.org/10.2788/101376>.
- Cucurachi, S., Sala, S., Laurent, A., Heijungs, R., 2014. Building and characterizing regional and global emission inventories of toxic pollutants. *Environ. Sci. Technol.* 48, 5674–5682. <https://doi.org/10.1021/es405798x>.
- Crenna, E., Secchi, M., Benini, L., Sala, S., 2019. Global environmental impacts: data sources and methodological choices for calculating normalisation factors. *Int. J. Life Cycle Assess.* 1–27. <https://doi.org/10.1007/s11367-018-1534-9>.
- Deblonde, T., Cossu-Leguille, C., Hartemann, P., 2011. Emerging pollutants in wastewater: a review of the literature. *Int. J. Hyg. Environ. Health* 214, 442–448. <https://doi.org/10.1016/j.ijheh.2011.08.002>.
- Dijkman, T.J., Birkved, M., Hauschild, M.Z., 2012. PestLCI 2.0: a second generation model for estimating emissions of pesticides from arable land in LCA. *Int. J. Life Cycle Assess.* 17, 973–986. <https://doi.org/10.1007/s11367-012-0439-2>.
- Dijkman, T.J., Basset-Mens, C., Antón, A., Núñez, M., 2018. LCA of food and agriculture. In: Hauschild, M.Z., Rosenbaum, R.K., Olsen, S.I. (Eds.), *Life Cycle Assessment. Theory and Practice*. Springer International Publishing, pp. 723–754.
- EC, 1998. Protocol to the 1979 Convention on Long-Range Transboundary Air Pollution on Persistent Organic Pollutants.
- EC, 2001. Organic Contaminants in Sewage Sludge for Agricultural Use.
- EC, 2006a. Regulation (EC) No 1907/2006 of the European Parliament and of the Council on the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH). (doi:2004R0726 - v.7 of 05.06.2013).
- EC, 2009b. Regulation (EC) No 166/2006 of the European Parliament and of the Council of 18 January 2006 concerning the establishment of a European pollutant release and transfer register and amending council directives 91/689/EEC and 96/61/EC.
- EC, 2007. Eurostat Statistical Books: The Use of Plant Protection Products in the European Union - Data 1992–2003, 2007. Brussels.
- EC, 2008. Directive 2008/105/EC of the European Parliament and of the Council of 16 December 2008 on environmental quality standards in the field of water policy. <http://eur-lex.europa.eu/legal-content/EN/TXT/?uri=celex:32008L0105>.
- EC, 2009a. Regulation (EC) No 1107/2009 of the European Parliament and of the Council of 21 October 2009 Concerning the Placing of Plant Protection Products on the Market and Repealing Council Directives 79/117/EEC and 91/414/EEC.
- EC, 2009b. Regulation (EC) No 1223/2009 of the European Parliament and of the Council of 30 November 2009 on Cosmetic Products.
- EC, 2010a. Communication from the Commission - Europe 2020: A Strategy for Smart, Sustainable and Inclusive Growth. (Brussels).
- EC, 2010b. Environmental, Economic and Social Impacts of the Use of Sewage Sludge on Land. Final Report - Part III: Project Interim Reports.
- EC, 2011. Communication from the Commission to the European Parliament, the Council, the European Economic and Social Committee and the Committee of the Regions. A Resource-Efficient Europe – Flagship Initiative under the Europe 2020 Strategy. Brussels.
- EC, 2012. Regulation (EU) no 528/2012 of the European Parliament and of the Council of 22 May 2012 concerning the making available on the market and use of biocidal products.
- EC, 2015. Communication from the Commission to the European Parliament, the Council, the European Economic and Social Committee and the Committee of the Regions - Closing the loop - An EU action plan for the Circular Economy COM(2015)/0614 final. (Brussels).
- EC, 2016. Communication from the Commission to the European Parliament, the Council, the European Economic and Social Committee and the Committee of the Regions - Next Steps for a Sustainable European Future: European Action for Sustainability COM(2016) 739 final. (Strasbourg).
- Ecobichon, D.J., 2001. Pesticide use in developing countries. *Toxicology* 160, 27–33. [https://doi.org/10.1016/S0300-483X\(00\)00452-2](https://doi.org/10.1016/S0300-483X(00)00452-2).
- EEA, 2013. The European pollutant release and transfer register (E-PRTR), member states reporting under article 7 of regulation (EC) no 166/2006 - version 5. <https://www.eea.europa.eu/data-and-maps/data/member-states-reporting-art-7-under-the-european-pollutant-release-and-transfer-register-e-prtr-regulation-7> (last accessed in 2013).
- EEA, 2016. Waterbase - emissions. <https://www.eea.europa.eu/data-and-maps/data/waterbase-emissions-5>, Accessed date: 21 September 2017.
- EEA, 2017. The European pollutant release and transfer register (E-PRTR), member states reporting under article 7 of regulation (EC) no 166/2006 - version 11. <https://www.eea.europa.eu/data-and-maps/data/member-states-reporting-art-7-under-the-european-pollutant-release-and-transfer-register-e-prtr-regulation-14>, Accessed date: 25 August 2017.
- EMEP/CEIP, 2017. Officially reported emission data. http://www.ceip.at/ms/ceip_home1/ceip_home/webdab_emepdatabase/reported_emissiondata/, Accessed date: 19 September 2017.
- EU, 2013. Decision No 1386/2013/EU of the European Parliament and of the Council of 20 November 2013 on a General Union Environment Action Programme to 2020 “Living Well, within the Limits of our Planet” (Text with EEA Relevance).
- European Chemical Agency, 2018. REACH registration results. <https://echa.europa.eu/reach-registrations-since-2008>, Accessed date: 27 June 2018.
- Eurostat, 2017a. National accounts aggregates by industry (up to NACE A*64) [nama_10_a64]. http://appsso.eurostat.ec.europa.eu/nui/show.do?dataset=nama_10_a64&lang=en, Accessed date: 10 December 2017.
- Eurostat, 2017b. Population connected to wastewater treatment plants [env_ww_con]. http://appsso.eurostat.ec.europa.eu/nui/show.do?dataset=env_ww_con&lang=en, Accessed date: 10 December 2017.
- Eurostat, 2017c. Population change - demographic balance and crude rates at national level [demo_gind]. http://appsso.eurostat.ec.europa.eu/nui/show.do?dataset=demo_gind, Accessed date: 10 June 2017.
- Eurostat, 2017d. Sewage sludge production and disposal [env_ww_spd]. http://appsso.eurostat.ec.europa.eu/nui/show.do?dataset=env_ww_spd&lang=en, Accessed date: 18 October 2017.
- Eurostat, 2017e. Crop statistics by NUTS 2 regions (from 2000 onwards) [agr_r_acs]. <http://ec.europa.eu/eurostat>, Accessed date: 19 October 2017.
- Fantke, P., Juraske, R., Antón, A., Friedrich, R., Joliet, O., 2011. Dynamic multicrop model to characterize impacts of pesticides in food. *Environ. Sci. Technol.* 45, 8842–8849. <https://doi.org/10.1021/es201989d>.
- Fiedler, H., 2007. National PCDD/PCDF release inventories under the Stockholm convention on persistent organic pollutants. *Chemosphere* 67, S96–S108. <https://doi.org/10.1016/j.chemosphere.2006.05.093>.
- Geelen, L.M.J., Huijbregts, M.A.J., den Hollander, H., Ragas, A.M.J., van Jaarsveld, H.A., de Zwart, D., 2009. Confronting environmental pressure, environmental quality and

- human health impact indicators of priority air emissions. *Atmos. Environ.* 43, 1613–1621. <https://doi.org/10.1016/j.atmosenv.2008.12.002>.
- Hamscher, G., Pawelzick, H.T., Höper, H., Nau, H., 2005. Different behavior of tetracyclines and sulfonamides in sandy soils after repeated fertilisation with liquid manure. *Environ. Toxicol. Chem.* 24, 861–868.
- Hauschild, M.Z., Rosenbaum, R.K., Olsen, S.I. (Eds.), 2018. *Life Cycle Assessment. Theory and Practice*. Springer International Publishing. <https://doi.org/10.1016/B978-0-12-801775-3.00024-X>.
- Hodges, J.E.N., Holmes, C.M., Vamshi, R., Mao, D., Price, O.R., 2012. Estimating chemical emissions from home and personal care products in China. *Environ. Pollut.* 165, 199–207. <https://doi.org/10.1016/j.envpol.2011.11.009>.
- Hodges, J.E.N., Vamshi, R., Holmes, C., Rowson, M., Miah, T., Price, O.R., 2014. Combining high-resolution gross domestic product data with home and personal care product market research data to generate a subnational emission inventory for Asia. *Integr. Environ. Assess. Manag.* 10, 237–246. <https://doi.org/10.1002/ieam.1476>.
- Kenessey, Z., 1987. The primary, secondary, tertiary and quaternary sectors of the economy. *Rev. Income Wealth* 359–385. <https://doi.org/10.1111/j.1475-4991.1987.tb00680.x>.
- Kuczyński, T., Dämmgen, U., Webb, J., Myczko, A., 2005. *Emissions from European Agriculture*. Wageningen Universiteit (Wageningen University).
- Laurent, A., Hauschild, M.Z., 2014. Impacts of NMVOC emissions on human health in European countries for 2000–2010: use of sector-specific substance profiles. *Atmos. Environ.* 85, 247–255. <https://doi.org/10.1016/j.atmosenv.2013.11.060>.
- Laurent, A., Owsianiak, M., 2017. Potentials and limitations of footprints for gauging environmental sustainability. *Curr. Opin. Environ. Sustain.* 25, 20–27. <https://doi.org/10.1016/j.cosust.2017.04.003>.
- Laurent, A., Lautier, A., Rosenbaum, R.K., Olsen, S.I., Hauschild, M.Z., 2011a. Normalization references for Europe and North America for application with USEtox™ characterization factors. *Int. J. Life Cycle Assess.* 16, 728–738. <https://doi.org/10.1007/s11367-011-0285-7> LCIA.
- Laurent, A., Olsen, S.I., Hauschild, M.Z., 2011b. Normalization in EDIP97 and EDIP2003: updated European inventory for 2004 and guidance towards a consistent use in practice. *Int. J. Life Cycle Assess.* 16, 401–409. <https://doi.org/10.1007/s11367-011-0278-6>.
- Lautier, A., Rosenbaum, R.K., Margni, M., Bare, J., Roy, P.-O., Deschênes, L., 2010. Development of normalization factors for Canada and the United States and comparison with European factors. *Sci. Total Environ.* 409, 33–42. <https://doi.org/10.1016/j.scitotenv.2010.09.016>.
- Leclerc, A., Laurent, A., 2017. Framework for estimating toxic releases from the application of manure on agricultural soil: national release inventories for heavy metals in 2000–2014. *Sci. Total Environ.* 590–591. <https://doi.org/10.1016/j.scitotenv.2017.01.117>.
- Lim, S.R., Lam, C.W., Schoenung, J.M., 2010. Quantity-based and toxicity-based evaluation of the U.S. toxics release inventory. *J. Hazard. Mater.* 178, 49–56. <https://doi.org/10.1016/j.jhazmat.2010.01.041>.
- Manfred, C., Haider, S., Poupas, S., Mareckova, K., Seuss, K., Windhofer, G., Zechmeister, A., Fischer, C., Ryberg, M., Hofbauer, P., Milunov, M., Seyring, N., 2012. *Three Years of Implementation of the E-PRTR: Supporting Study for the European Commission*.
- De Marchi, S., Hamilton, J.T., 2006. Assessing the accuracy of self-reported data: an evaluation of the toxics release inventory. *J. Risk Uncertain.* 32, 57–76. <https://doi.org/10.1007/s10797-006-6666-3>.
- Mareckova, K., Wankmueller, R., Moosmann, L., Pinterits, M., Tista, M., 2013. *Methodology Report. Review of Emission Data Reported under the LRTAP Convention and NEC Directive, Technical Report CEIP*.
- Miège, C., Choubert, J.M., Ribeiro, L., Eusèbe, M., Coquery, M., 2009. Fate of pharmaceuticals and personal care products in wastewater treatment plants - conception of a database and first results. *Environ. Pollut.* 157, 1721–1726. <https://doi.org/10.1016/j.envpol.2008.11.045>.
- Muntean, M., Janssens-Maenhout, G., Song, S., Giang, A., Selin, N.E., Zhong, H., Zhao, Y., Olivier, J.G.J., Guizzardi, D., Crippa, M., Schaaf, E., Dentener, F., 2018. Evaluating EDGARv4.4tox2 speciated mercury emissions ex-post scenarios and their impacts on modelled global and regional wet deposition patterns. *Atmos. Environ.* 184, 56–68. <https://doi.org/10.1016/j.atmosenv.2018.04.017>.
- Nriagu, J.O., Pacyna, J.M., 1988. Quantitative assessment of worldwide contamination of air, water and soils by trace elements. *Nature* 333, 134–139. <https://doi.org/10.1038/333134a0>.
- OECD, 2017. *Wastewater treatment (% population connected)*. http://stats.oecd.org/index.aspx?DataSetCode=water_treat, Accessed date: 10 December 2017.
- OECD, 2018. *Pollutant release and transfer register*. <http://www.oecd.org/chemicalsafety/pollutant-release-transfer-register/>, Accessed date: 16 August 2018.
- Pacyna, J.M., Pacyna, E.G., 2001. An assessment of global and regional emissions of trace metals to the atmosphere from anthropogenic sources worldwide. *Environ. Rev.* 9, 269–298. <https://doi.org/10.1139/er-9-4-269>.
- Pacyna, E.G., Pacyna, J.M., Fudala, J., Strzelecka-Jastrzab, E., Hlawiczka, S., Panasiuk, D., Nitter, S., Pregger, T., Pfeiffer, H., Friedrich, R., 2007. Current and future emissions of selected heavy metals to the atmosphere from anthropogenic sources in Europe. *Atmos. Environ.* 41, 8557–8566. <https://doi.org/10.1016/j.atmosenv.2007.07.040>.
- Persson, L., Karlsson-Vinkhuyzen, S., Lai, A., Persson, Å., Fick, S., 2017. The globally harmonized system of classification and labelling of chemicals-explaining the legal implementation gap. *Sustainability* 9. <https://doi.org/10.3390/su9122176>.
- Persson, L., Arvidsson, R., Berglund, M., Cederberg, C., Finnveden, G., Palm, V., Sörme, L., Schmidt, S., Wood, R., 2019. Indicators for national consumption-based accounting of chemicals. *J. Clean. Prod.* 215, 1–12. <https://doi.org/10.1016/j.jclepro.2018.12.294>.
- Poupa, S., Mareckova, K., Seuss, K., Van Der Maas, W., Kuenen, J., Dröge, R., Dore, C., 2014. *Methodologies for Identifying Incomplete Reporting of E-PRTR Emission Data with Releases to Air, ETC/ACM Technical Paper*.
- Quass, U., Fermann, M., Bröker, G., 2000. *The European Dioxin Emission Inventory Stage II Volume 3 Assessment of Dioxin Emissions until 2005*.
- Rosenbaum, R.K., Anton, A., Bengoa, X., Björn, A., Brain, R., Bulle, C., Cosme, N., Dijkman, T.J., Fantke, P., Felix, M., Geoghegan, T.S., Gottesbüren, B., Hammer, C., Humbert, S., Joliet, O., Juraske, R., Lewis, F., Maxime, D., Nemecek, T., Payet, J., Räsänen, K., Roux, P., Schau, E.M., Sourisseau, S., van Zelm, R., von Streit, B., Wallman, M., 2015. The Glasgow consensus on the delineation between pesticide emission inventory and impact assessment for LCA. *Int. J. Life Cycle Assess.* 20, 765–776. <https://doi.org/10.1007/s11367-015-0871-1>.
- Sala, S., Goralczyk, M., 2013. Chemical footprint: a methodological framework for bridging life cycle assessment and planetary boundaries for chemical pollution. *Integr. Environ. Assess. Manag.* 9, 623–632. <https://doi.org/10.1002/ieam.1471>.
- Sala, S., Benini, L., Ponsioen, T., Laurent, A., van Zelm, R., Stam, G., Goralczyk, M., Pant, R., 2014. Methodology for Building LCA-Compliant National Inventories of Emissions and Resource Extraction. <https://doi.org/10.2788/18238>.
- Sala, S., Benini, L., Mancini, L., Pant, R., 2015. Integrated assessment of environmental impact of Europe in 2010: data sources and extrapolation strategies for calculating normalisation factors. *Int. J. Life Cycle Assess.* 20, 1568–1585. <https://doi.org/10.1007/s11367-015-0958-8>.
- Sleeswijk, A.W., van Oers, L., Guinée, J., Struijs, J., Huijbregts, M., 2008. Normalisation in product life cycle assessment: an LCA of the global and European economic systems in the year 2000. *Sci. Total Environ.* 390, 227–240. <https://doi.org/10.1016/j.scitotenv.2007.09.040>.
- Steinmann, Z.J.N., Schipper, A.M., Stadler, K., Wood, R., de Koning, A., Tukker, A., Huijbregts, M.A.J., 2018. Headline environmental indicators revisited with the global multi-regional input-output database EXIOBASE. *J. Ind. Ecol.* 22, 565–573. <https://doi.org/10.1111/jiec.12694>.
- The World Bank, 2017. *Urban population*. <http://databank.worldbank.org/data/reports.aspx?source=2&series=SP.URB.TOTL.IN.ZS&country=>, Accessed date: 12 October 2017.
- Tobiszewski, M., Namieśnik, J., 2012. PAH diagnostic ratios for the identification of pollution emission sources. *Environ. Pollut.* 162, 110–119. <https://doi.org/10.1016/j.envpol.2011.10.025>.
- Tukker, A., Giljum, S., Wood, R., 2018. Recent progress in assessment of resource efficiency and environmental impacts embodied in trade: an introduction to this special issue. *J. Ind. Ecol.* 22, 489–501. <https://doi.org/10.1111/jiec.12736>.
- Ulrich, B., 2016. *E-PRTR Informal Data Review 2016 Joint Meeting of IEEG and E-PRTR Expert Group on Industrial Emissions Reporting*.
- UN, 2010. *The 1998 Protocol on Persistent Organic Pollutants, Including the Amendments Adopted by the Parties on 18 December 2009*.
- UNECE, 2016. *PRTR Global map*. <https://prtr.unece.org/prtr-global-map>, Accessed date: 16 August 2018.
- UNEP, 2011. *Decoupling natural resource use and environmental impacts from economic growth. In: A Report of the Working Group on Decoupling to the International Resource Panel*.
- UNEP, 2013. *Global mercury assessment. In: Glob. Mercur. Assess. 2013 Sources, Emiss. Releases Environ. Transp. vol. 3. pp. 1–258 (doi:DTI/1636/GE)*.
- UN-HABITAT, 2008. *Global Atlas of Excreta, Wastewater Sludge, and Biosolids Management: Moving Forward the Sustainable and Welcome Uses of a Global Resource*.
- UNITAR, 2018. *PRTR platform*. <http://prtr.unitar.org/site/home>, Accessed date: 16 August 2018.
- United Nations, 2015. *Resolution Adopted by the General Assembly on 25 September 2015 70/1. Transforming our World: The 2030 Agenda for Sustainable Development*.
- Verlicchi, P., Al Aukidy, M., Zambello, E., 2012. Occurrence of pharmaceutical compounds in urban wastewater: removal, mass load and environmental risk after a secondary treatment-a review. *Sci. Total Environ.* 429, 123–155. <https://doi.org/10.1016/j.scitotenv.2012.04.028>.
- WHO, 2017. *Guidelines for Drinking-water Quality: Fourth Edition Incorporating the First Addendum*. WHO, Geneva. <https://doi.org/10.2105/AJPH.17.2.162-b>.
- Zhang, Y., Tao, S., 2009. Global atmospheric emission inventory of polycyclic aromatic hydrocarbons (PAHs) for 2004. *Atm. Env.* 43 (4), 812–819. <https://doi.org/10.1016/j.atmosenv.2008.10.050>.
- Zijp, M.C., Posthuma, L., Van De Meent, D., 2014. Definition and applications of a versatile chemical pollution footprint methodology. *Environ. Sci. Technol.* 48, 10588–10597. <https://doi.org/10.1021/es500629f>.